Accelerator Mass Spectrometry – an alternative to decay counting to enable the study of previously inaccessible nuclear reactions.

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UCRL-PRES-233586

National Ignition Facility – Nuclear Astrophysics Workshop, August 27- 30 2007.

The success of astrophysical models depends strongly on accuracy of nuclear data.

Nucleosynthesis gives us one of the very important fingerprints of nature: elemental and isotopic abundances.

Despite progress made with astrophysical models, we still have much to learn about the physics of nuclear reactions that lead to these distributions.

One essential key to this request is the precise knowledge of reaction and production cross-sections. This will lead to:

- **well established inputs to astrophysical models of nucleosynthesis.**
- **improved understanding of novae, X-ray burster, and supernovae.**
- **verification of nuclear model calculations.**
- **understanding of the forces responsible for nuclear interactions.**

Well-established data for many important nuclear reactions are lacking – in part due to deficiencies of current detection methodologies.

New detection tools and schemes are needed to study previously inaccessible nuclear reactions.

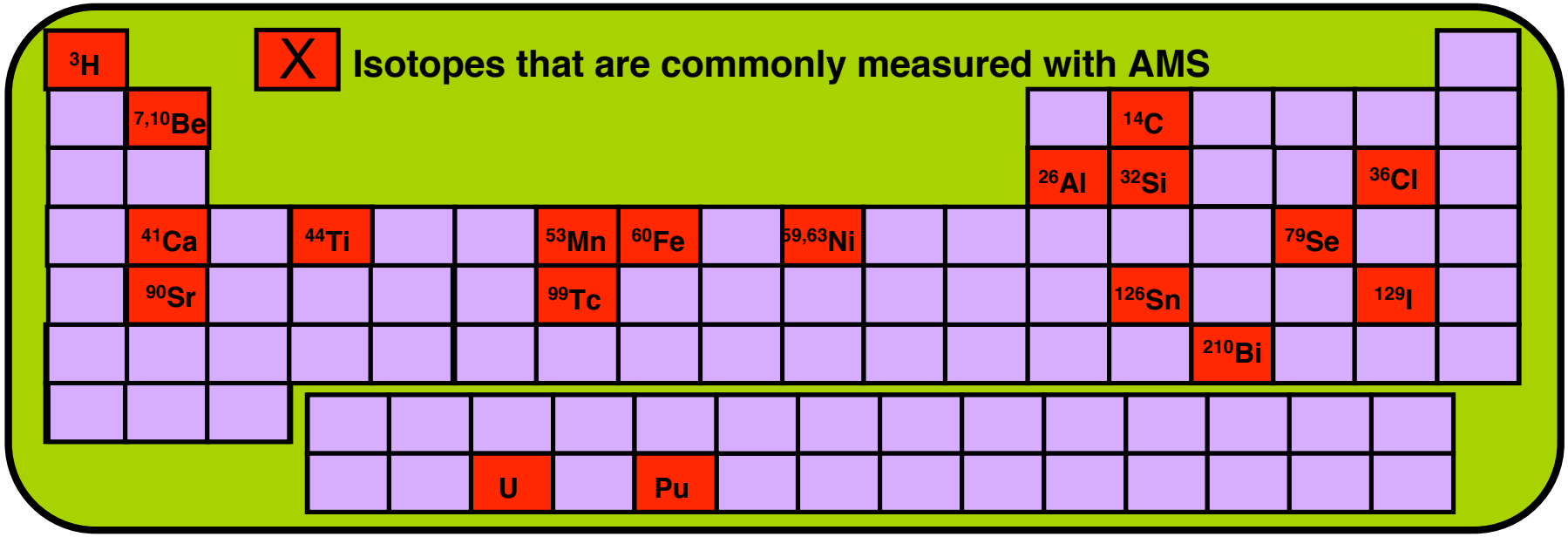
Nuclear reaction and production cross-sections are typically measured using time of flight and decay counting schemes.

Cross-section data for many nuclides are lacking as decay counting techniques have difficulty quantifying nuclear reactions that:

- **have small cross-sections.**
- **are performed on a paucity of starting material.**
- **possess unfavorable decay schemes:**
  - **produce long lived radionuclides.**
  - **decay via low energy electron or x-ray emission.**

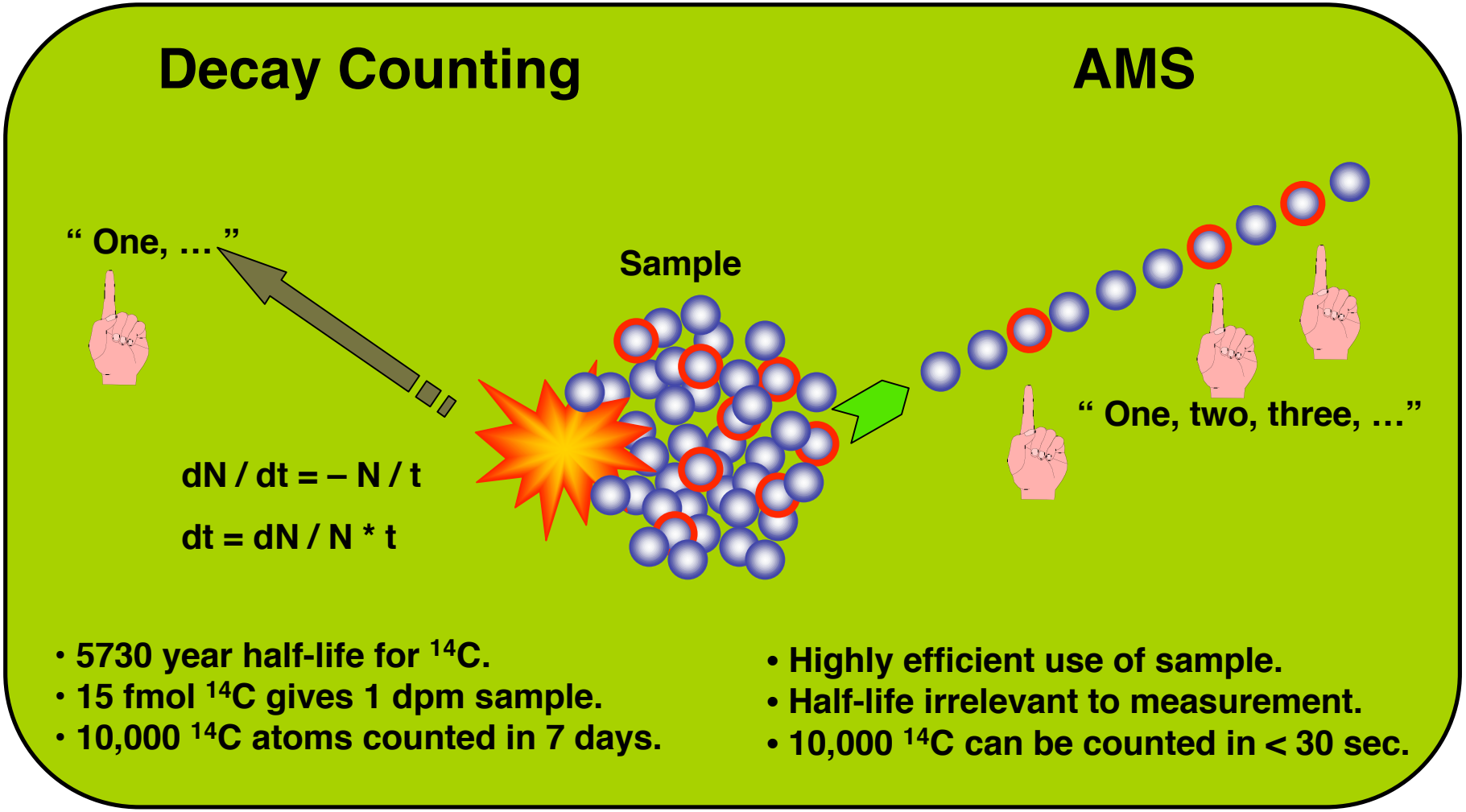
Sensitive techniques, such as AMS, that count individual atoms offer an alternative means to probe reactions that cannot be precisely quantified via decay counting.

Accelerator Mass Spectrometry (AMS) is routinely used to precisely quantify rare, long-lived isotopes.

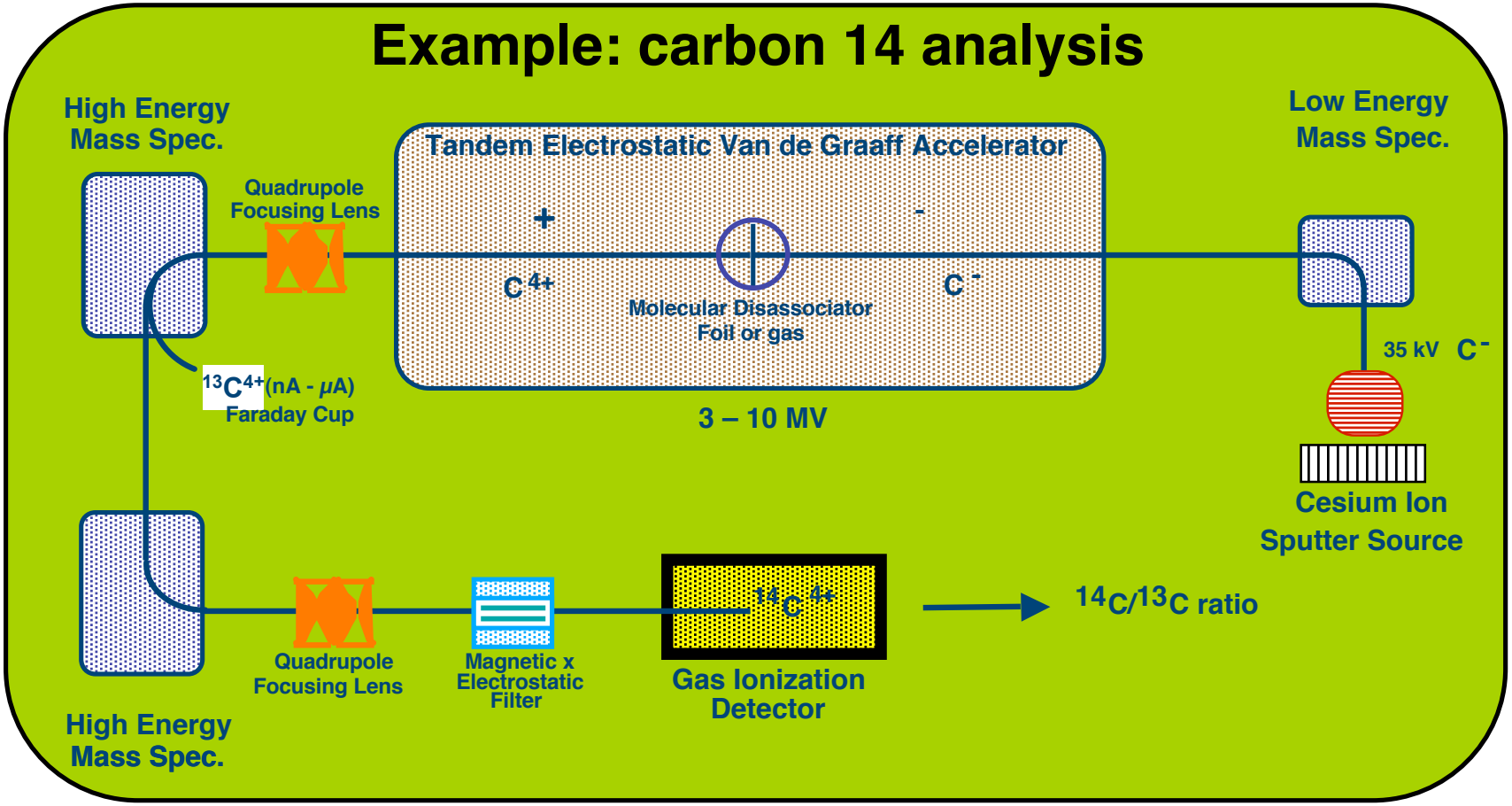


- AMS counts atoms of an individual isotope.
- Isotope ratio dynamic range ~ 10^{-9} to 10^{-18}.
- Detection limits ~ 10^{3} to 10^{6} atoms.
- Efficient destruction of molecular isobars.
- Amenable to the addition of matrix carriers during sample preparation.
- Attomole quantitation in mg sample sizes.

AMS counts atoms, not decays.



AMS measures isotope ratios: Separates ions based on mass, charge & energy.



Ions energies are MeV rather than KeV as in conventional mass spectrometry enabling efficient destruction of molecular isobars in the accelerator.

AMS makes efficient use of the sample and has very low background interference.

Measurement efficiency ranges from ~1% for heavy elements to ~ 20% for carbon and enables isotope detection limits ranging from ~10^4 to 10^6 atoms.

Destruction of molecular isobars results in low background interference.

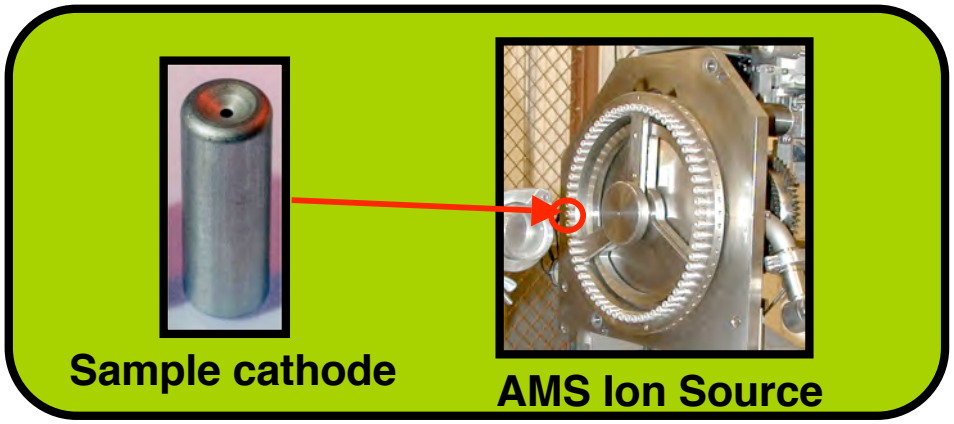
Isotope	Background ratio
^{14}C	1×10^{-15}
^{3}H	4×10^{-15}
^{10}Be	5×10^{-17}
^{26}Al	2×10^{-15}
^{36}Cl	3×10^{-15}
^{41}Ca	1×10^{-13}
^{129}I	2×10^{-14}
Actinides	$< 5 \times 10^5$ atoms

These features enable quantification of low concentrations of an isotope and afford the addition of matrix carriers during sample preparation for more robust analysis.

AMS requires solid samples in an appropriate chemical/physical form for robust analysis.

Sample preparation is performed using well understood, established procedures.

1) Addition of appropriate carrier material or matrix spike (if necessary).
2) Homogenization of sample – typically via digestion or combustion.
3) Chemical or physical separation of element of interest (if necessary).
4) Conversion of sample to solid – typically via precipitation (including precipitation onto a suitable matrix carrier), reduction or drying.
5) Solid material packed into sample cathode for AMS analysis.

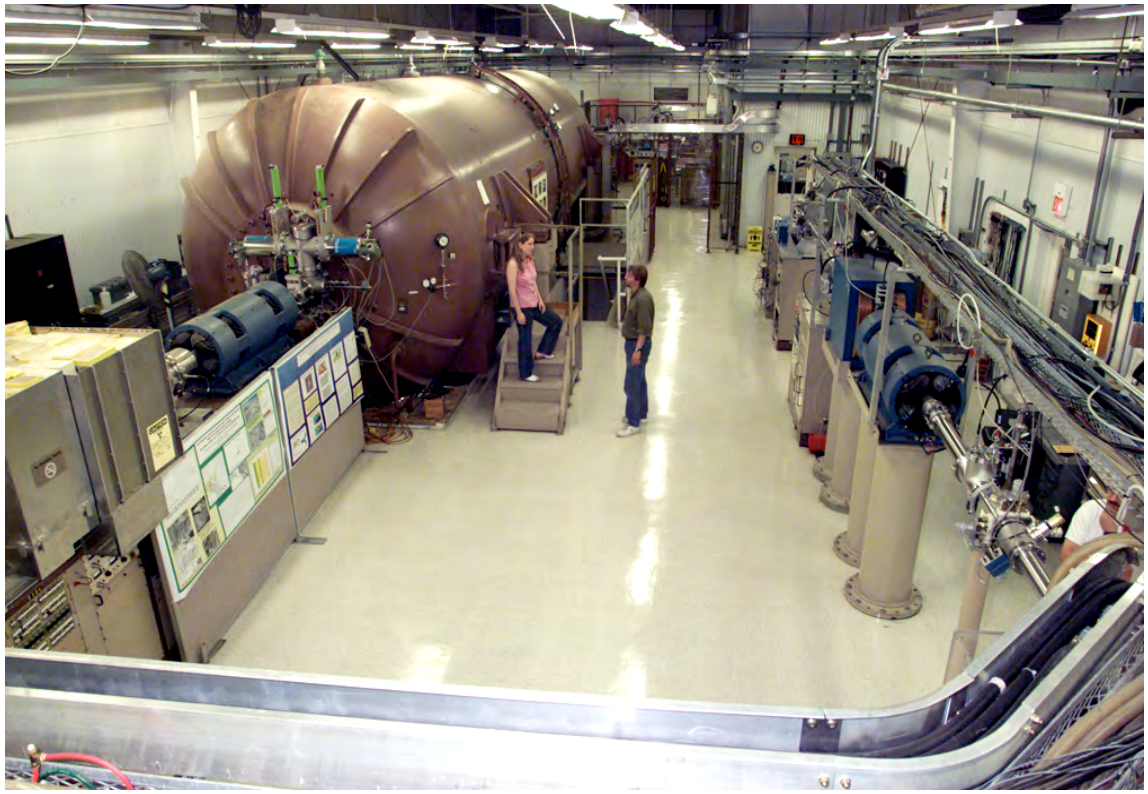


Owing to destruction of molecular isobars sample preparation for AMS is frequently simpler than for other atom counting techniques.

LLNL's CAMS is the most versatile and productive AMS facility in the world.

Standard measurement of ^{3}H, ^{7},^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{63}Ni, ^{129}I, ^{233,234,36}U, $^{238–244}$Pu.

Approximately 25,000 AMS analyses performed per year.



Contributes to LLNL programs in carbon sequestration, climate change, geo- and bio-sciences, and national security.

AMS has already been used for several cross-section and half live measurements.

Cross-section measurements

(most performed at astrophysically relevant energies).

- $^{13}C(n; \gamma)\ ^{14}C$
- $^{14}N(^{16}O; \alpha)\ ^{26}Al$
- $^{16}O(n; x)\ ^{14}C$
- $^{25}Mg(p; \gamma)\ ^{26}Al$
- $^{36}Ar(n;p)\ ^{36}Cl$
- $^{40}Ar(n; x)\ ^{36}Cl$
- $^{40}Ca(\alpha; \gamma)\ ^{44}Ti$
- $^{40}Ca(n; \gamma)\ ^{41}Ca$
- $^{58}Ni(n: \gamma)\ ^{59}Ni$
- $^{62}Ni(n; \gamma)\ ^{63}Ni$
- $^{78}Se(n; \gamma)\ ^{79}Se$
- $^{209}Bi(n; \gamma)\ ^{210}Bi$

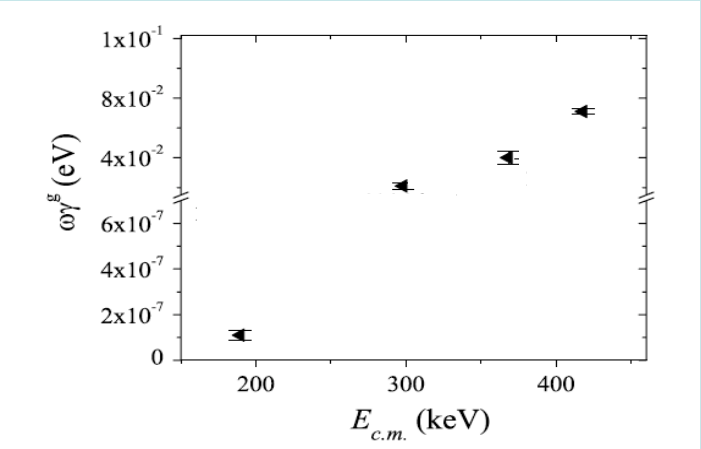
Half life determinations.

- ^{32}Si
- ^{44}Ti
- ^{41}Ca
- ^{60}Fe
- ^{126}Sn
- ^{209}Bi

$^{25}Mg(p; \gamma)\ ^{26}Al$

Strength of resonances for $^{25}Mg(p; \gamma)$ reaction for the formation of ^{26}Al in its ground state

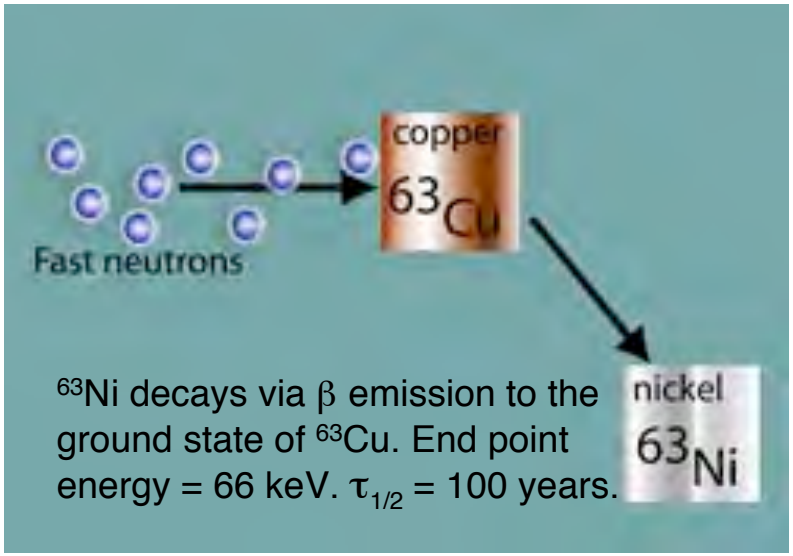
Phys Rev C 74 025802 (2006)



The high efficiency of AMS and very low background interference enabled precise measurement on samples containing very small numbers of atoms of interest.

Measurement of ^{63}Ni for retrospective determination of neutron flux from the Hiroshima atomic bomb.

- **Survivors of the atomic bomb are used to estimate cancer risks due to radiation exposure.**
- **Doubts have been raised over the accuracy of calculated neutron doses.**



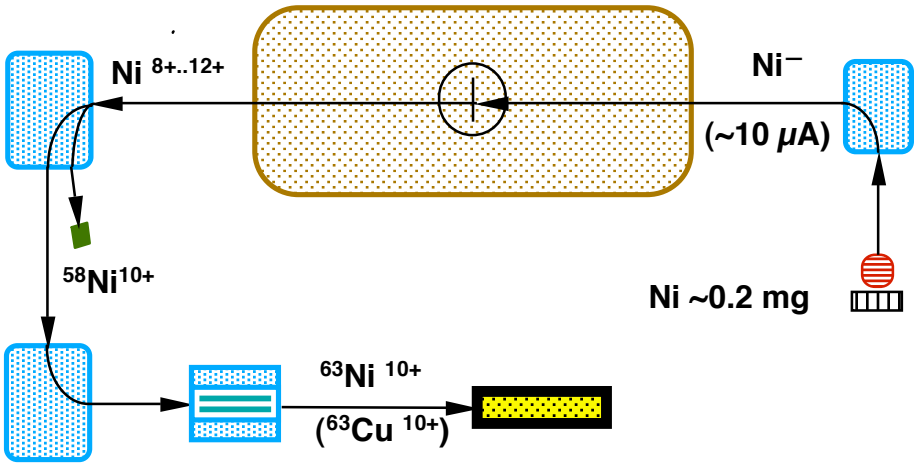
Trace amounts (parts per 10^{18} Cu) of ^{63}Ni were formed by the passage of fast neutrons through copper materials.

Copper materials (lightning conductors, rain gutters, roofing) of known location were analyzed.

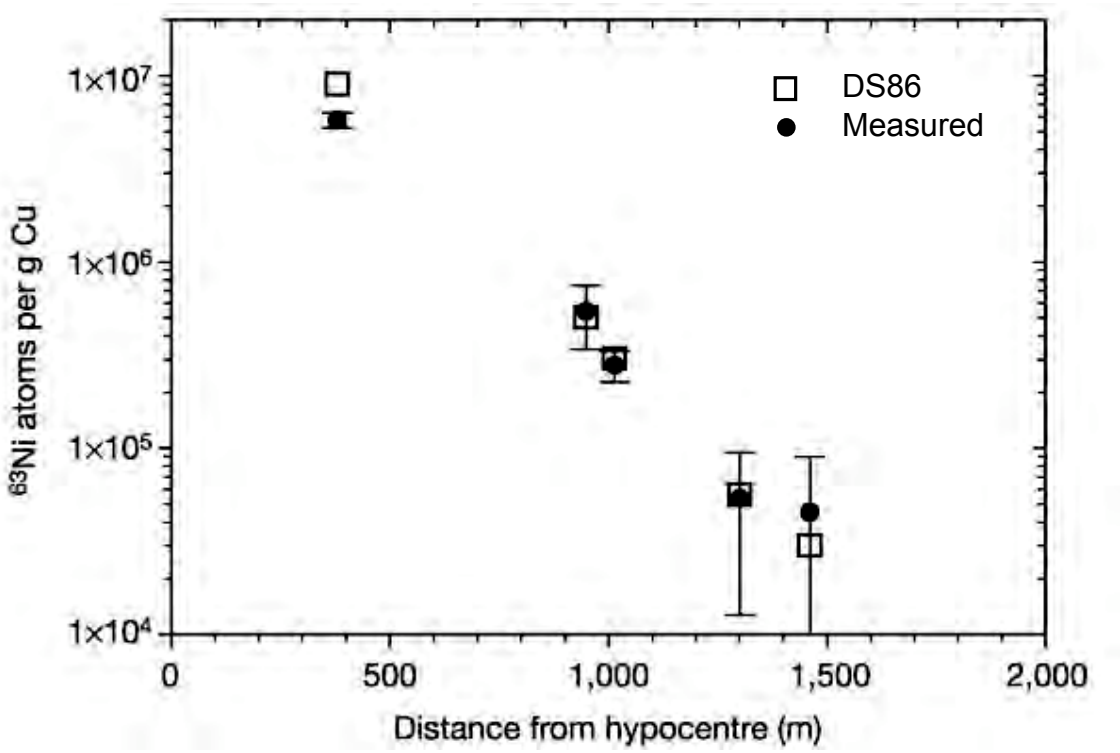
Sample preparation presented a major technical challenge.

- ^{63}Cu represents a difficult isobaric interference for mass spectrometry measurements of ^{63}Ni.
- Electrochemical separation resulted in a 10^{12} reduction of Cu.
- Ni + 4CO → $Ni(CO)_4$
  Thermally decomposes to Ni in ion source.

AMS systems at CAMS and Munich were used to measure ^{63}Ni contents.



^{63}Ni-AMS measurements validated neutron dose estimates.

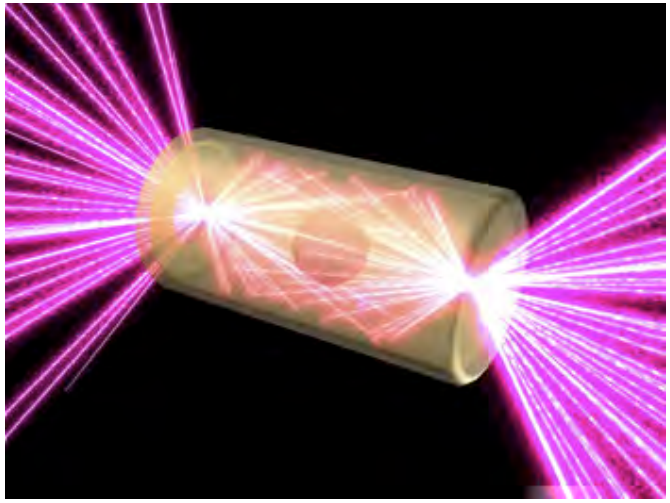


***Nature* 424: 539 (2003)**

Measured levels of ^{63}Ni are in line with sample-specific computer modeling calculations based on the DS86 dosimetry system at distances relevant to survivors, so concerns about large dose discrepancies can be discounted.

AMS has potential for quantifying reaction products from NIF based experiments.

High-energy-density regimes accessible though NIF experiments will yield new insight into the origin of our universe.



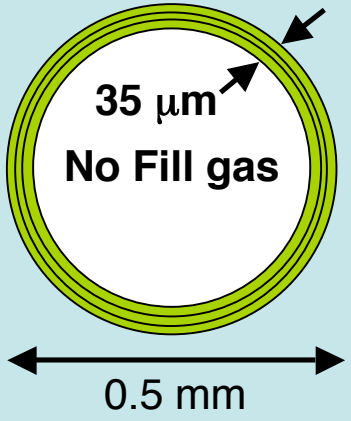
Experiments will be conducted on mm sized shells that will likely produce ~ 10^4 to 10^{10} reaction products per shot.

The high efficiency of AMS coupled with very low background interference, high sensitivity and relative ease of sample preparation should enable precise measurement on samples containing small numbers of radioactive or stable atoms.

Example 1: Measurement of ^{10}Be for determination of background levels in "virgin NIF" capsules.

^{9}Be(n;γ) ^{10}Be could serve as a potential neutron flux monitor.

Be(Cu) Capsules
~ 40 μg Be, 5 μg Cu



Sample Preparation

Each capsule digested in aqua regia (3 HCl : 1 HNO_3)
~10 μg aliquots from each digest used for AMS
100 μg of Be carrier in dilute nitric acid added
NH_4OH added and $Be(OH)_2$ precipitated
Precipitate dried and ignited to produce BeO

Capsule #	^{10}Be/ ^{9}Be ratio in shell
1	(1.13 +/- 0.25) E-14
2	(1.04+/- 0.26) E-14

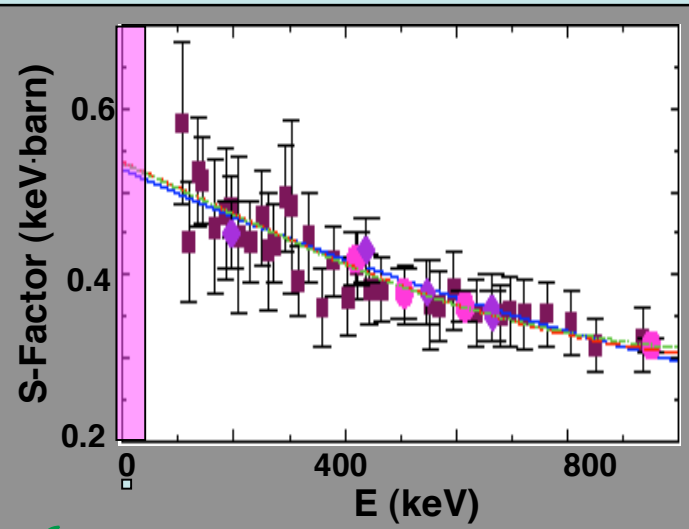
Ratios are consistent with those obtained from commercial sources of Be.

Data indicate ~μg quantities of Be could be analyzed with better than 10% precision.

Example 2: Measuring $^{3}He(^{4}He;\gamma)\ ^{7}Be$ with NIF

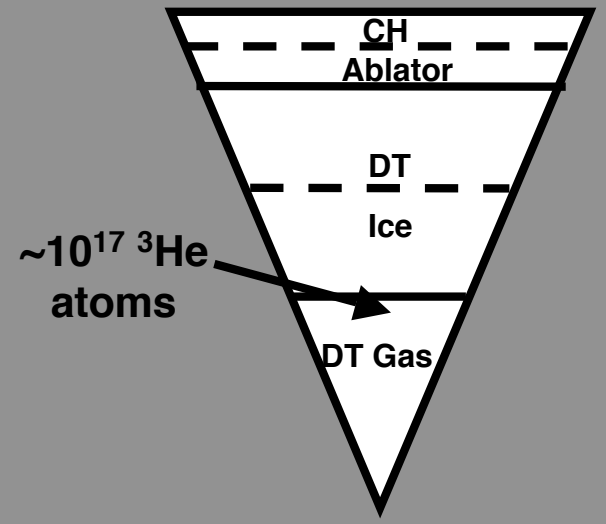
Thermonuclear reactions can be observed in 'passive' NIF implosions or as by products of the temperature runaway in d + t burn – but measurement challenging!

Accelerator-Based Experiments



- ✓ Mono-energetic
- ✗ Low event rate (few events/month)
- ✗ Not performed at relevant energies

NIF-Based Experiments



- ✓ High Count rate ($3x10^5$ atoms/shot)
- ✓ Energy window is better

AMS can perform Be isotope measurements on as few as 10^3 atoms – hence ^{7}Be measurement via AMS may be tractable given sufficient sample recovery.

Efficient sample collection methods will be needed for NIF to exploit off-line measurement techniques.

The number of reaction products produced per shot may approach detection limits of counting techniques - necessitates efficient sample collection methods.

Some thoughts and questions:

Gaseous products may be tractable to collect at high efficiencies with strategically placed pumps.

Particulate and debris collectors currently recover a small fraction of the sample.

Particulate aerosol collectors exist that have high collection efficiencies for 0.1 to 10 mm diameter particles at atmospheric pressure. Can these be made to operate with high collection efficiencies at lower pressures?

To what level will legacy reaction products from previous shots and are there ways to mitigate against such effects?